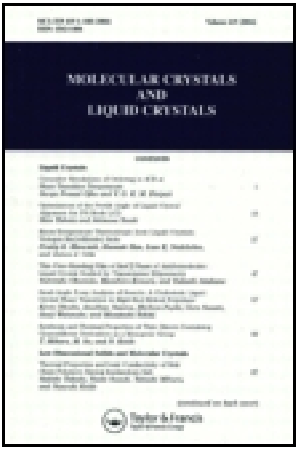

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Dynamics of Magnetization for a System (S=3) with Strong Uniaxial Magnetocrystalline Anisotropy

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We study the classical and the quantum mechanical dynamics of the magnetization for a model system (S=3) with strong uniaxial magnetocystalline anisotropy. The characteristic features of the hysteresis loop and the dependence of the staircase structure on the field sweep rate are discussed and a suitable sweep rate for observing many quantum steps is proposed.

Keywords: staircase structure; hysteresis; non adiabatic transition; quantum tunneling; time dependence of magnetization; uniaxial magnet

INTRODUCTION

Recently, considerable effort has been devoted to the study of magnetic quantum tunnelling (MQT)[1-12]. Especially, manganese cluster (Mn_{12}-Ac) has been the subject of much experimental and theoretical work. Magnetization hysteresis is a familiar macroscopic phenomenon in the behavior of magnetic materials. But until recent years no one had ever seen quantum steps in a hysteresis loop. The Mn_{12}-Ac cluster[13] has the net spin S=10 and is oriented along the c-axis of the tetragonal unit cells as a result of its strong uniaxial magnetocrytalline anisotoropy. A magnetic field applied along the c-axis makes one of the two anisotropy wells energetically metastable. Due to strong uniaxial magnetocrytalline anisotoropy[14], the system shows a clear magnetization hysteresis loop. In addition, the hysteresis loops have a distinct staircase structure[15-18]. Hysteresis is observed below blocking temperature and the relaxation time obeys an Arrhenius law at first stage as the temperature decreases. At lower temperatures, however, the relaxation time does not freeze out, but behaves independently of temperature. It has been interpreted as possible evidence of MQT of the spin in these high-spin molecules.

The following spin Hamiltonian has been proposed for this system in labeling the easy-magnetization axis of the crystal z[14, 16].

$$\hat{H} = -DS_z^2 - g\mu_B \boldsymbol{S}\cdot\boldsymbol{h} \equiv -DS_z^2 - \boldsymbol{S}\cdot\boldsymbol{H} \quad (1)$$

$$\text{or} \quad \hat{H} = -DS_z^2 - g\mu_B S_z h_z + \hat{H}' \equiv -DS_z^2 - S_z H_z + \hat{H}' , \quad (2)$$

where $\boldsymbol{h}$ is the external magnetic field, $g\mu_B$ is the gyromagnetic ratio of the electron and D is the uniaxial anisotropy energy constant. $\hat{H}'$ is a perturbation which does not commute with S_z.

We investigate the classical and the quantum mechanical dynamics of the magnetization for a model system (S=3) with strong uniaxial magnetocystalline anisotropy. We perform a Monte Carlo simulation for the classical model system using the hamiltonian (1), and show magnetization hysteresis loops. The behavior of hysteresis is examined with the dependence on parameters D and T (temperature). We also study the time dependence of the magnetization which is obtained from the solution of the time-dependent Schrödinger equation, where the hamiltonian (2) is adopted, and present the correlation between staircase structures and field sweep rates in the view of the Landau-Zener tunneling picture.

Theoretical Back Ground and Method

Let us consider a model system (S=3) described by Eq. (1) or (2). In the absence of an external magnetic field, the system has pair of degenerate potential wells. When an external field is applied in the positive z direction, the degeneracy is broken and the energy levels for m > 0 sink deeper, while those in the other well rise. When two levels on opposite sites of the potential barrier have the same energy, quantum tunneling between them becomes strongly enhanced, which is often called resonant tunneling.

Recently H. De Raedt, S. Miyashita and coworkers showed that the Landau-Zener tunneling picture correctly describes the time dependence of the magnetization of a uniaxial magnet on a slowly changing applied field[19-23]. In this study we use the same methodology and investigate the time dependence of the magnetization of quantum spin system with strong uniaxial magnetocrystalline anisotropy (S=3).

First we consider the classical system described by Eq. (1). Using a Monte Carlo simulation, we observed the statistical average of magnetization for 1000 sites imposing a field parallel to z axis. The applied filed H starts from 3.2, where the initial state is made to be the equilibrium state, decreases to -3.2 and returns to the initial value 3.2. In this process H is changed by 0.1 after 30 Monte Carlo steps proceed. The following three cases are adopted; A(D=0.7, T=1.0), B(D=0.7, T=0.5), and C(D=1.0, T=1.0), taking $k_B = 1$.

Next we study the quantum mechanical dynamics of the magnetization. Here transverse field Γ is introduced as a perturbation in Eq. (2) to observe the effect of a quantum fluctuation, and this perturbation term induces tunneling.

$$\hat{H} = -DS_z^2 - S_z H + S_x \Gamma \cdot \tag{3}$$

In Fig 1. we present the energy levels of this model as a function of the applied field H for D=1 and Γ=0. In the case of $\Gamma \neq 0$ each crossing point becomes avoided and near this point Landau-Zener transition probability p has some value and the quantum tunneling can occur. Making use of this picture, staircase structures will be observed. The time dependence of the magnetization is obtained from the solution of the time-dependent Schrödinger equation. The applied field is changed from H_0=6 to $-H_0$=-6 gradually at a constant field sweep rate c:

$$H(t) = H_0 - c\,t. \tag{4}$$

Assuming that t_u is the number of steps used in a simulation, c is defined as $2H_0 / t_u$. If Γ is not so large, the ground state for H=6 can be considered as the eigenstate of S_z, ie.,m=3. This point is chosen as the starting point in the following simulation and referred to s shown in Fig. 1. We observe the magnetization $<M_z(t)>$ and the correlation between the magnetization and the sweep rate. We take $\hbar=1$ as a unit of energy and fix $D=1$ and $\Gamma=0.5$.

Results and Discussion

Fig. 2 and 3 show the hysteresis loops for cases A and B, respectively, where M means the statistical average of the magnetization per site. A characteristic feature of these hysteresis curves is observed in the difference of the coercive fields H_c, which are the external fields necessary to return the magnetization to zero. In adopting lower temperature of case B, H_c is larger and the hysteresis becomes clear. Choosing larger D has the same effect. In fact, for case C the hysteresis loop is found to be very similar to case B. We actually confirmed that low temperature and large D are necessary conditions to show a clear hysteresis.

In the quantum mechanical treatment, we performed simulations for sweep

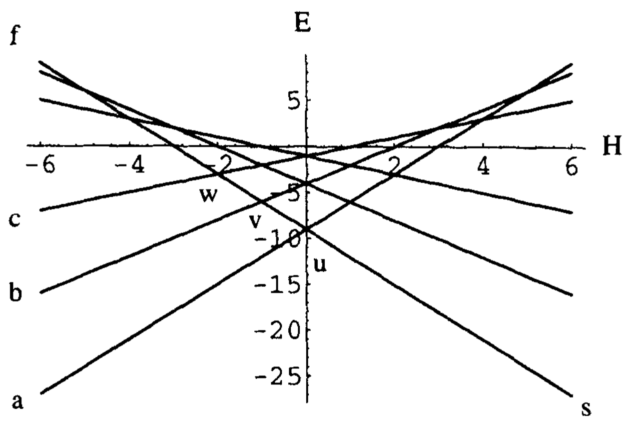

FIGURE 1 Energy levels of the model described by Eq. (3) as a function of the applied field for $D=1$ and $\Gamma=0$.

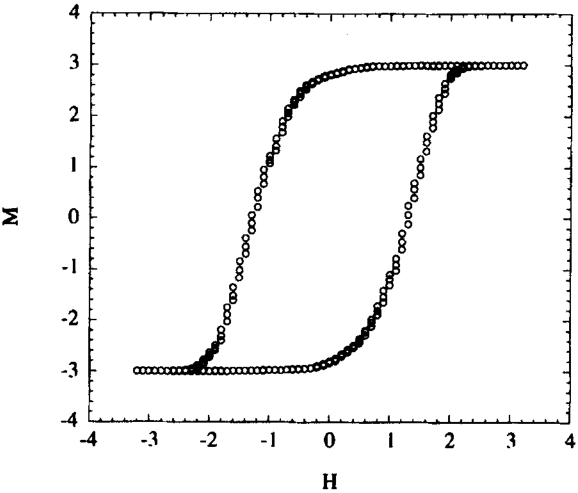

FIGURE 2 Hysteresis loop obtained by a Monte Carlo simulation for case A.

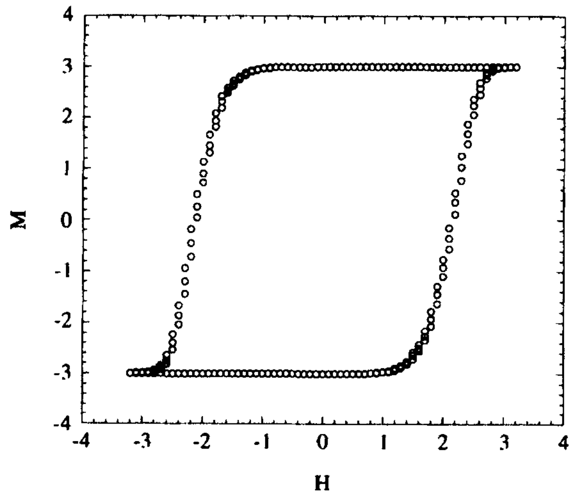

FIGURE 3 Hysteresis loop obtained by a Monte Carlo simulation for case B.

rates c=1.25x10^{-7}, 5.0x10^{-6}, 5.0x10^{-5}, 1.0x10^{-3}, and 1.0x10^{-2}. If we consider only two states m and m', according to the Landau-Zener theory, the probability p of changing the state from m to m' such as s ---->a throughout u, etc., shown in Fig. 1 is given by

$$p = 1 - \exp\left(-(\Delta E)^2 / d\right), \tag{5}$$

where ΔE is the energy splitting at the crossing point and d is proportional to the sweep rate c.

Figure 4 shows the staircase structure of magnetization for the sweep rate c=1.25x10^{-7}. Because of a very slow sweep rate, p is almost 1 at the first crossing point u. It is found from Fig. 4 that the state of this system changes adiabatically and the final state becomes the ground state for H=-6. For faster sweep rate non adiabatic transition begins to occur and another route s ----> u ----> v can be added besides the path s ----> u ----> a. In Fig. 5 two steps are appeared at H=0 and H=-1. The first step at H=0 means that the tunneling occurs in rather lower probability p. The residual probability corresponds to the path s ----> v ----> b. There exists little probability for the path s ----> v ----> w because <M> for H=-6 is almost -2. From Fig. 6 it is concluded that for c=5.0x10^{-5} one process is dominant: s ----> v ----> b. In Fig. 7 for c=1.0x10^{-3} there exist two steps, where s ----> v ----> b and s ----> w ----> c processes occur simultaneously. In the case of c=1.0x10^{-2} one process is dominant: s ----> w ----> c as shown in Fig. 8.

It is important to note that the sweep rate changes the tunneling probability and that the tunneling probability at each crossing point is generally different from one another even if the sweep rate is fixed. Therefore, various staircase structures could be possible in changing the external parameter. From above analysis, the tunneling probability becomes larger as its corresponding crossing point is located on lower H for a constant sweeping rate. All the possible quantum steps will be scarcely observed if the sweep rate is constant during a sweep, which is adopted throughout our study. Choosing the sweep rate suitably at each external filed H, where the rate has time dependence, this observation could become possible. By making the sweep rate slow at the beginning of a sweep and accelerating this rate with time or sweeping, more complex stair-

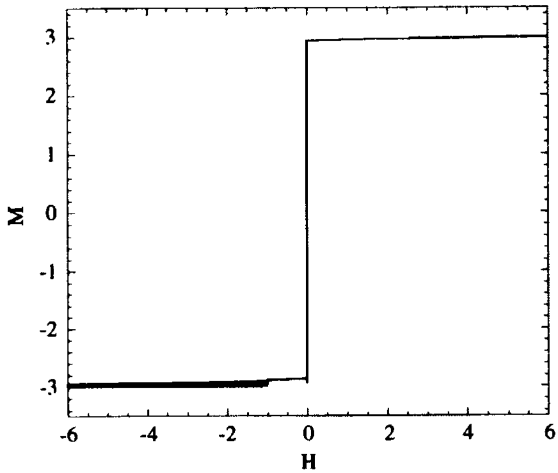

FIGURE 4 Staircase structure of magnetization for the sweep rate c=1.25x10^{-7}.

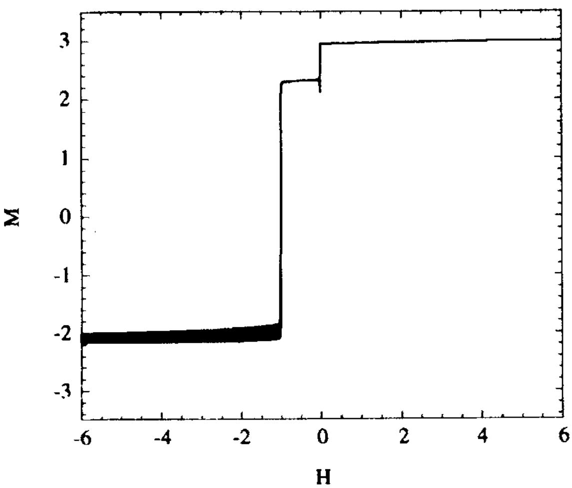

FIGURE 5 Staircase structure of magnetization for the sweep rate c= 5.0x10^{-6}.

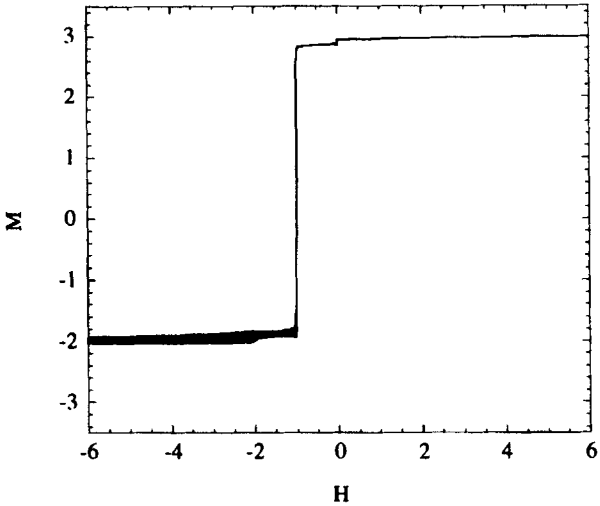

FIGURE 6 Staircase structure of magnetization for the sweep rate $c = 5.0\times10^{-5}$.

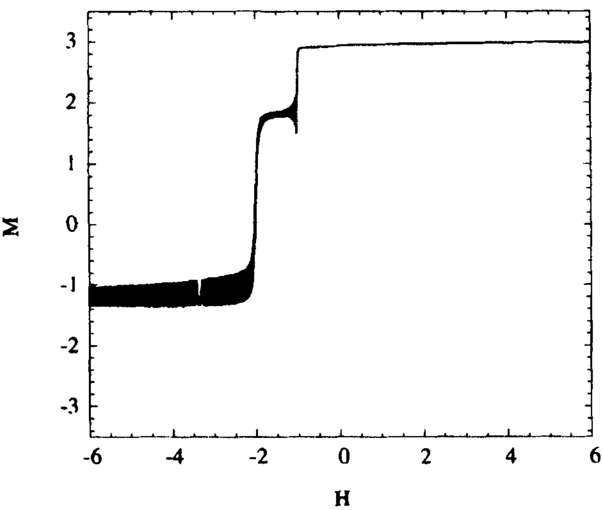

FIGURE 7 Staircase structure of magnetization for the sweep rate $c = 1.0\times10^{-3}$

case structures with many quantum steps could be obtained. For example the sweep rate should be changed as 5.0×10^{-6} ----> 1.0×10^{-3} ----> in this study.

We neglected the thermal noise effect in the quantum mechanical treatment and focussed on the qualitative discussion of MQT. A quantitative treatment and a study of thermal noise effect will be performed in the future.

Concluding Remarks

We confirmed the characteristic features of the hysteresis loop with the dependence on parameters *T* and *D* in a Monte Carlo simulation for a classical model (S=3). Low temperature and large *D* are necessary conditions to show a clear hysteresis. Staircase structures with many steps would be observed by choosing the sweep rate suitably at each external filed *H*, which comes from the quantum mechanical treatment. In reducing the applied magnetic field from maximum value, small sweep rate should be kept at first stage and the sweep should be accelerated gradually as the filed decreases.

Acknowledgments

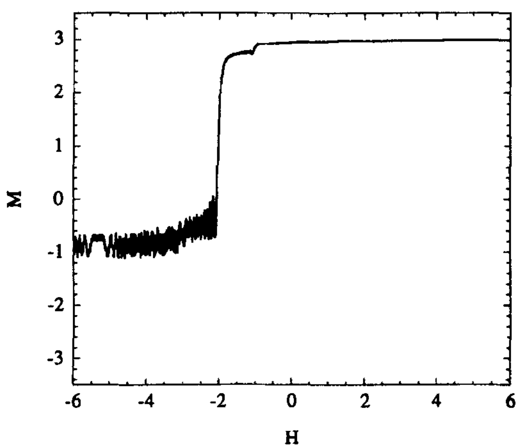

FIGURE 8 Staircase structure of magnetization for the sweep rate $c = 1.0 \times 10^{-2}$.

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